GEORGIA INSTITUTE OF TECHNOLOGY
OFFICE OF CONTRACT ADMINISTRATION
SPONSORED PROJECT INITIATION

Date: June 13, 1979

Project Title: Theoretical Investigation and Calculation of Rates of Certain Atomic and Molecular Processes Important in the Formation and Destruction of Excited Stable Rare Gas Molecules (or Excimers)

Project No: G-41-679 (Green card)

Project Director: Dr. M. R. Flannery

Sponsor: Department of Energy; Oak Ridge Operations; Oak Ridge, TN 37830

Agreement Period: From 10/1/78 Until ~~9/30/79~~ 11/30/79 (Contract Period)

Type Agreement: Contract No. DE-AS05-76DP40017, Mod. No. M004
(formerly Contract No. EY-76-S-05-5002)

Amount: $20,000 DOE Funds (G-41-679)
4,423 GIT Contribution (G-41-322)
5,071 (DOE unexpended funds from previous year)
$29,494 Total

Reports Required: Publication Preprints; Publication Reprints; Quarterly Progress Reports; Final Report

Sponsor Contact Person (s):

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Oak Ridge, TN 37830

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NOTE: FOLLOW-ON PROJECT TO G-41-665.

Defense Priority Rating: N/A

Assigned to: Physics (School/Laboratory)

COPIES TO:

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- Division Chief (EES)
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- Project Code (GTRI)
- Other

GEORGIA INSTITUTE OF TECHNOLOGY
OFFICE OF CONTRACT ADMINISTRATION

SPONSORED PROJECT TERMINATION

Date: April 9, 1981

Project Title: Theoretical Investigation and Calculation of Rates of Certain Atomic & Molecular Processes Important in the Formation & Destruction of Excited Stable Rare Gas Molecules

Project No: G-41-609

Project Director: Dr. M. R. Flannery

Sponsor: DOE - Oak Ridge Operation

Effective Termination Date: 11/30/79

Clearance of Accounting Charges: 11/30/79

Grant/Contract Closeout Actions Remaining:

_ Final Invoice and Closing Documents
_ Final Fiscal Report
_x Final Report of Inventions
_x Govt. Property Inventory & Related Certificate
_ Classified Material Certificate
_ Other _____

Assigned to: Physics (School/Laboratory)

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CA–4 (1/79)

DOE Form IR-427
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U.S. DEPARTMENT OF ENERGY

UNIVERSITY-TYPE CONTRACTOR AND GRANTEE RECOMMENDATIONS FOR DISPOSITION OF SCIENTIFIC AND TECHNICAL DOCUMENT

See Instructions on Reverse Side

1. DOE Report No. DOE/DP/76DP40017

2. Contract No. DE-AS05-76DP40017

3. Title THEORETICAL INVESTIGATION AND CALCULATION OF CERTAIN ATOMIC AND MOLECULAR PROCESSES IMPORTANT IN THE FORMATION AND DESTRUCTION OF EXCITED STABLE GAS MOLECULES (OR EXCIMERS)

4. Type of Document ("X" one)

☒ a. Scientific and technical report

☐ b. Conference paper:

Title of conference _____

Date of conference _____

Exact location of conference _____

Sponsoring organization _____

☐ c. Other (Specify Thesis, Translations, etc.)

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Are there any patent related objections to the release of this information product? If so, state these objections.

8. Submitted by — Name and Position (Please print or type)

M. R. Flannery, Professor

Organization
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332

Signature /

Date April 2, 1981

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9. Patent Clearance ("x" one)

☐ a. DOE patent clearance has been granted by responsible DOE patent group.

☐ b. Report has been sent to responsible DOE patent group for clearance.

☐ c. Patent clearance not required.

DOE/DP/76DP40017

THEORETICAL INVESTIGATION AND CALCULATION OF CERTAIN ATOMIC AND MOLECULAR PROCESSES IMPORTANT IN THE FORMATION AND DESTRUCTION OF EXCITED STABLE GAS MOLECULES (OR EXCIMERS)

Final Report

M. R. Flannery

School of Physics
Georgia Institute of Technology
Atlanta, Georgia

October 1, 1978 - November 30, 1979

PREPARED FOR THE DIVISION OF LASER FUSION
U. S. DEPARTMENT OF ENERGY
UNDER CONTRACT NO. DE-AS05-76DP40017
(formerly EY-76-S-05-5002)

Table of Contents

1. Title and Abstract

Title: Theoretical Investigation and Calculation of Certain Atomic and Molecular Processes Important in the Formation and Destruction of Excited Stable Gas Molecules (or Excimers).

Abstract: The work performed during the contract period 1 October 1978 - 30 November 1979 is fully documented and is as follows:

(a) Atomic and Molecular Collision Processes in Rare-Gas-Halide Lasers and Rare-Gas Excimer Lasers.

(b) Semiquantal Treatment of Excited-Atom-Excited-Atom Collisions.

(c) Systematic Trends in the Inelastic Cross Sections and Form Factors for $n\ell \rightarrow n'\ell'$ Direct Collisional Transitions.

(d) Multistate Semiclassical Orbital Treatment of Li^+-H_2 and H^+-H_2 Collisions.

(e) Elastic Scattering and Rotational Excitation in Ion-Molecule Collisions. II. Li^+-H_2 and H^+-H_2 Collisions.

2. Work Performed and Results Obtained

The following topics were initiated and completed since the previous Technical Progress Report ORO-EY-76-S-05-5002/78 covering the period October 1, 1977 - September 30, 1978:

(A) Atomic and Molecular Collision Processes in Rare-Gas-Halide Lasers and Rare-Gas Excimer Lasers.

The key cycles of atomic and molecular collision processes contributing to the formation and quenching of the excited molecular states in exciplex (such as KrF^*) laser systems are delineated and discussed.

(B) Semiquantal Treatment of Excited-Atom-Excited-Atom Collisions.

The semiquantal treatment of collision processes such as $H(n\ell) + H(n_o\ell_o)$ $H(n'\ell') + H^+ + e$ is described. The associated cross sections display systematic trends in the distribution over final angular momentum states in keeping with those previously predicted for $e-H(n\ell)$ inelastic collisions. Deexcitation between close-neighboring levels n and n' of the projectile (with simultaneous ionization of the target) is generally more rapid than the endothermic processes involving the reverse transition in the projectile and, as n is increased, becomes quite competitive with those processes associated with no internal-energy change in the projectile, particularly at the lower impact energies. The cross sections, with appropriate statistical weights, for forward and reverse transitions in the projectile tend to the same high-energy limit.

(C) Systematic Trends in the Inelastic Cross Sections and Form Factors for $n\ell \rightarrow n'\ell'$ Direct Collisional Transitions.

Certain theoretical predictions are presented for the preferential population of final states with angular momentum ℓ' in collisions involving an initially excited atom. Varying ℓ', we find that the maxima of both the inelastic form factors and cross sections for the $n\ell \rightarrow n'\ell'$ transitions in hydrogen,

induced by collision with electrons and heavy particles, in general, oscillate on a background which rises as ℓ' is increased, until they both attain a pronounced peak at a unique value ℓ'_{max} which is strongly dependent on only the initial principal quantum number n and which is fairly insensitive to changes in ℓ and n'. An expression for ℓ'_{max} is derived. For $\ell' > \ell'_{max}$, the form factors and associated cross sections exhibit a dramatic decline, resulting in negligible population of those states. The predictions differ from those suggested by the Bethe high-energy asymptotic limit which favors dipole transitions, and assume significance in situations where excited states are important as in laser modeling, astrophysical and fusion plasmas, and in laboratory studies of excited Rydberg states. For heavy-particle ($n\ell \rightarrow n'\ell'$) collisional transitions the additional undulations which appear in the cross sections over a wide energy range are predicted and explained.

(D) Multistate Semiclassical Orbital Treatment of Li^+-H_2 and H^+-H_2 Collisions.

A semiclassical orbital description of elastic scattering and of rotational excitation in ion-molecule collisions is presented. Specific account is taken of interference effects between the classical trajectories that contribute to a given scattering angle, and of rainbow scattering. Excellent agreement with full quantal differential cross sections is obtained.

(E) Elastic Scattering and Rotational Excitation in Ion-Molecule Collisions. II. Li^+-H_2 and H^+-H_2 Collisions.

A general semiclassical treatment of elastic scattering and of rotational excitation in ion-molecule collisions is presented. When the orbits associated with the different channels corresponding to the internal modes do not differ significantly, simplification occurs and the internal degrees of freedom can then be coupled to the relative motion via the introduction of an optical

potential (which in turn depends on the transition amplitudes). Total energy is consequently conserved. An expression is derived for the inelastic scattering amplitude which acknowledges various interference effects and possible rainbow scattering. With all phase-information supressed, the procedure, when compared with the full quantum-mechanical results, reproduces the background elastic and inelastic scattering in Li^+-H_2 and in H^+-H_2 collisions. Restoration of the phases, particularly of the eikonal or action phases associated with the different classical paths that contribute to a specified scattering angle, produces the interference oscillations present in the differential cross section for scattering angles less than the rainbow angle. The method, when compared with the full quantal procedure, is remarkably efficient and accurate.

3. Refereed Papers Published Under Contract.

Since the initiation of contract DE-AS05-76DP40017 (formerly EY-76-S-05-5002) on October 1, 1976 until its completion on September 30, 1979 the following work has been completed, written up for publication and published:

1. "Molecular Charge Transfer Reactions of Tritium," with T. F. Moran, K. J. McCann and D. L. Albritton, J. Chem. Phys. 65, 3172-80 (1976).
2. "Formation of Vibrationally Excited Neutral Molecules in Charge Transfer Reactions," with T. F. Moran, J. Phys. B (Atom. Molec. Phys.) 9, 1509-12 (1976).
3. "The Effect of Reactant Vibrational State on Differential Charge Transfer Cross Sections for T_2^+ ($X^2\ \Sigma^+_g, v_o'$) + T_2($X\ ^1\Sigma^+_g, v''$) Collisions," with K. J. McCann and T. F. Moran, Chem. Phys. Letts. 39, 374-78 (1976).
4. "Ionic Recombination," in Atomic Processes and Applications, ed. P. B. Burke and B. L. Moiseiwitsch (Amsterdam, North Holland) pp. 407-466 (1976).
5. "Cross Sections for Ionization of Metastable Rare Gas Atoms (Ne^*, A^*, Kr^*, Xe^*) and of Metastable N_2 and CO^* by Electron Impact," with D. Ton-That, Phys. Rev. A 16, 517-26 (1977).
6. "Photoionization of Vibrationally Excited Molecular Hydrogen," with H. Tai, Phys. Rev. A 16, 1124-1135 (1977).
7. "Photoionization of Metastable Rare-Gas Atoms (He^*, Ne^*, Ar^*, Kr^*, Xe^*)," with K. J. McCann, Appl. Phys. Letts. 31, 599-601 (1977).
8. "Cross Sections for the Photoionization of X_2($X\ ^1\Sigma^+_g$, v_i=0-14) with the formation of H_2^+($X\ ^2\Sigma^+_g$, v_f=0-18), Vibrational Overlaps and R^n-Centroids (n=1,2) for the Associated $H_2(v_i) \rightarrow H^+_2(v_f)$ Vibrational Transitions," with H. Tai and D. L. Albritton, At. Data and Nucl. Data Tables 20, 563-585 (1977).
9. "Ionic Recombination of Rare-Gas Atomic Ions X^+ with F^- in a Dense Gas X," with T. P. Yang, Appl. Phys. Letts. 32, 327-329 (1978).

10. "Ionic Recombination of Rare-Gas Molecular Ions X_2^+ with F^- in a Dense Gas X," with T. P. Yang, Appl. Phys. Letts. 32, 356-357 (1978).

11. "Three-Body Ion-Ion Recombination in Mercury-Halide Lasers," Chem. Phys. Letts. 56, 143-148 (1978).

12. "Ionic Recombination of Kr^+ and Kr_2^+ with F^- in Dense Buffer Rare Gases," with T. P. Yang, Appl. Phys. Letts. 33, 574-576 (1978).

13. "Elastic Scattering and Rotational Excitation in Ion-Molecule Collisions II, Li^+-H_2 and H^+-H_2 Collisions," with K. J. McCann, J. Chem. Phys. 69, 5275 (1978).

14. "Systematic Trends in the Inelastic Cross Sections and Form-Factors for $n\ell \rightarrow n'\ell'$ Direct Collisional Transitions," with K. J. McCann, J. Phys. B 12, 427 (1979).

15. "Multistate Semiclassical Orbital Treatment of Li^+-H_2 and H^+-H_2 Collisions," with K. J. McCann, Chem. Phys. Letts. 60, 523-527 (1979).

16. "Semiquantal Treatment of Excited Atom-Excited Atom Collisions," with K. J. McCann, Phys. Rev. A 19, 2206-2213 (1979).

17. "Atomic and Molecular Collision Processes in Rare-Halide Lasers and Rare Gas-Excimer Lasers," int. J. Quant. Chem.: Quant. Chem. Symp. 13, 501-529 (1979).